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54 Process and apparatus for treating sulphur dioxide.

57 A sulphur dioxide-containing gas, for example, obtained by a process using ammonia to remove SO_x and NO_x, is treated by the use of a reaction apparatus in which the gas is reduced and in which the composition of the gas is controlled by oxidation, to convert the composition of the gas into one suitable for the Claus reaction.

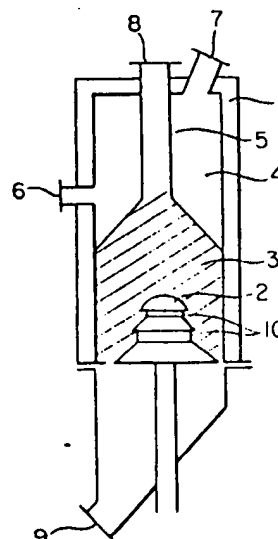


FIG. 1

EP 0 217 567 A2

"PROCESS AND APPARATUS FOR TREATING SULPHUR DIOXIDE"

BACKGROUND OF THE INVENTION

5 1. Field of the Invention

This invention relates to the treatment of sulphur dioxide and particularly to the production of sulphur from gas containing sulphur dioxide. More particularly, it is concerned with treating a gas
10 containing sulphur dioxide to convert it into a composition suitable for the Claus reaction process.

The production of elementary sulphur by a dry process comprising reducing sulphur dioxide sulphur and hydrogen sulphide and then obtaining elementary
15 sulphur using a condenser and a Claus reactor has hitherto been carried out by bringing the gas containing sulphur dioxide (SO_2) into contact with a carbonaceous material such as coal, reducing and directly obtaining elementary sulphur, or by reducing
20 SO_2 in the gas containing it to form hydrogen sulphide (H_2S) and then subjecting the resulting mixture gases of H_2S and SO_2 in a proportion of 2 : 1 to the Claus reaction to obtain sulphur.

In these processes, there have been proposed
25 various methods of preparing raw material gases for the Claus reaction. One method comprises contacting a gas containing SO_2 with a carbonaceous material to reduce part of the SO_2 and thereby adjusting the ratio of H_2S and SO_2 to 2 : 1 (Japanese Patent
30 Application OPI (Kokai) No. 167107/1982). Another method comprises dividing a gas containing SO_2 into two parts with a volume ratio of 2 : 1, reducing wholly the part with the larger volume to H_2S and mixing this with the other part to adjust the ratio
35 of H_2S and SO_2 to 2 : 1 (Japanese Patent Application OPI (Kokai) No. 32307/1981). A third

method comprises reducing part or all of the SO_2 in the gas containing SO_2 to obtain a gas containing an excessive amount of H_2S and then subjecting the gas to partial combustion in a combustion furnace provided according to the method employed in the desulphurization process for the ordinary petroleum refining process to adjust the ratio of hydrogen sulphide and sulphur dioxide to 2 : 1

The above described methods have various disadvantages:

It is difficult to maintain constant a ratio of hydrogen sulphide and sulphur dioxide by controlling the reducing reaction. Moreover, a by-pass is required. A gas containing sulphur dioxide in a high concentration obtained by an SO_x and NO_x removal process using ammonia, contains ammonia in a proportion of up to 1.0 % in some cases. Furthermore, after the reduction of the gas containing SO_2 a part of the gas is then fed to a combustion furnace in which the temperature is maintained by the combustion of liquefied petroleum gas. The conversion into sulphur dioxide is achieved by feeding air in an excessive amount, thereby controlling the ratio of H_2S and SO_2 . It is necessary to install a combustion furnace for controlling the composition: the amount of the gas to be processed is increased because of the use of the excessive air, thus lowering the concentration of the reaction components and the efficiency of the Claus reaction.

SUMMARY OF THE INVENTION

5 It is an object of the present invention to provide an improved process for the production or extraction of sulphur from a gas containing SO_2 .

10 It is a further or alternative object of the present invention to provide an improved process for treating a gas containing SO_2 to convert the composition thereof into that suitable for the Claus reaction.

15 According to one aspect the invention provides a process for treating a gas containing sulphur dioxide, comprising subjecting the gas to a reducing reaction and controlling by oxidation the composition of the gas to render the composition suitable for the Claus reaction.

20 The invention also provides apparatus for treating a gas containing sulphur dioxide, comprising means for subjecting the gas to a reducing reaction and means for controlling the composition of the reduced gas by oxidation.

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BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a schematic view of one embodiment of an apparatus for treating a gas containing SO_2 .

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Figure 2 is a schematic view of another embodiment of an apparatus for treating a gas containing SO_2 .

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Figure 3 is a flow diagram to illustrate a process for producing sulphur from a gas containing SO_2 by the Claus reaction.

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Figure 4 is a schematic view of a further embodiment of an apparatus for treating a gas containing SO_2 .

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DETAILED DESCRIPTION

5 The present invention is based on treating a gas containing SO_2 by reduction and controlling the composition of the gas by oxidation, preferably performed in adjacent reactors or in the same reactor.

10 In the present invention, carbonaceous materials such as coal and coke and reducing gases such as carbon monoxide and hydrogen can be used as a reducing agent and air or other oxygen-containing gases can be used as an oxidizing agent.

15 In the apparatus shown in Figure 1, a gas containing SO_2 is fed to a treatment apparatus 1 from an inlet 10, brought into contact with a carbonaceous material at a temperature between 600 to 1000°C in a reducing part 3 filled with carbonaceous material. About 80 to 100% of SO_2 in the inlet gas is reduced to sulphur, hydrogen sulphide, carbonyl sulphide, etc. The reducing part 20 3 is preferably a moving bed reactor filled with a carbonaceous material supplied from an inlet 8 by way of a chute 5. The resultant ash and non-fired carbonaceous material are discharged from an exhaust port 9. 2 represents a grate for discharging the carbonaceous material.

25 The temperature of the reducing part 3 can be maintained by feeding air or an oxygen-containing gas separately or together with the raw inlet gas so as to burn a part of the reducing agent.

30 The thus reduced gas is partially oxidized by air fed from an air inlet 6 in a composition controlling part 4 (combustion chamber) provided above the reducing reaction part 3 preferably at a temperature of 500 to 900°C to form sulphur oxide, wherebv the ratio of hydrogen sulphide and sulphur 35 dioxide, i.e. the value of $(\text{H}_2\text{S} + \text{COS} +$

0.5CS₂)/SO₂ is adjusted to 1.50 to 2.50,
preferably 1.90 to 2.10, suitable for the Claus
reaction, and then discharged from a gas outlet 7.
The rate of the oxidation reaction can be freely
5 controlled by changing the quantity of air supplied.

Figure 2 shows another embodiment of an
apparatus for treating a sulphur dioxide-containing
gas according to the present invention, in which a
composition controlling reactor 4 is adjacent to a
10 reducing reactor 3, the other parts corresponding to
those described with reference to Figure 1.

When a combination of a carbonaceous material
and a reducing gas is employed, the apparatus of
Figure 1 or Figure 2 can be used as described. The
15 reducing gas may be supplied together with the gas
containing SO₂ from the gas inlet 10. If a reducing
gas is used alone, the temperature should be
maintained by burning either a part of the reducing
gas or fuel supplied for the purpose.

20 One example of a process for treating gas
containing SO₂ is shown in Figure 3. A gas 26
containing SO₂ is fed to an apparatus 21 in which
the gas is subjected to a partial or complete
reduction of SO₂ then oxidized again to provide a
25 composition suitable for the Claus reaction,
subjected to removal of by-produced sulphur in a
condenser (not shown) and then fed to a Claus reactor
22. The condenser may be omitted. An air supply 23
provides air for the composition controlling
30 reaction. The rate of air flow is being controlled
by valve 24 according to the ratio of H₂S and the
other necessary components to SO₂ in a tail gas 29
of the Claus reaction. The ratio is determined by a
meter 25.

35 Figure 4 shows a further embodiment of an
apparatus for treating gas containing SO₂, the

apparatus comprising gas inlets 10 and 11 for raw material gas and reducing gas respectively, packing 12 such as broken bricks, a gas injection pipe 13, a fire grate 14, a combustion chamber 15 and a burner 16.

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There are in general several advantages arising from the preferred practice of the invention. Since reduction and the control of composition by oxidization can be performed in a single reactor or in a series of adjacent reactors, a combustion furnace for controlling the composition is not needed. Therefore, a supply of air for the combustion of liquefied petroleum gas, etc, is not required, so there is no increase of the process gas and the concentration of the reaction components can be higher. Furthermore, the contact time with a catalyst in a Claus reactor can so be lengthened to improve the efficiency of recovery of sulphur. The tail or exhaust gas of the Claus reaction is decreased. Moreover, a temperature of 500 to 800 °C can readily be maintained by the combustion of combustible components in the reduced gas without using fuels such as liquefied petroleum gas. Furthermore, it is possible to control the furnace temperature in the composition controlling part and the composition of the gas for the Claus reaction by controlling only the supply of air, control of the operation and composition is easy. In addition, whereas a gas containing SO₂ and obtained by a dry process which removes SO_x and NO_x using ammonia contains up to 1% ammonia in some cases, according to the present invention gas can be treated such that the ammonia is substantially decomposed to give a low concentration, e.g. 500 to 1500 ppm, and a subsequent Claus reaction is scarcely affected. Finally, a gas containing SO₂ and obtained by a process of

removing SO_x or SO_x and NO_x contains sulphur trioxide. However sulphur trioxide can be reduced by treating the whole gas according to the present invention and thus corrosion due to formation of sulphuric acid can be prevented.

The following example is given in order to illustrate the present invention without limiting the same.

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Example 1

Using an apparatus as shown in Figure 1 and coke as a reducing agent, a raw material gas having a composition of 20-25% H_2O , 20-25% SO_2 , 25-35% N_2 and 20-30% CO_2 was treated to produce sulphur according to the process as shown in Figure 3. The gas flow rate was 40-50 m^3/hr , the temperature in the reducing part was of 700 to 800°C, maintained by feeding air from the gas inlet 10 and burning the reducing agent. The temperature in the past was 600 - 700°C and thus the gas for the Claus reaction was controlled to give an H_2S etc./ SO_2 ratio of 1.95 to 2.05. Sulphur was prepared with a yield of 90 to 95% and a purity of elementary sulphur of at least 99.99%.

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Example 2

The procedure of Example 1 was repeated except that a gas having a composition of 40-45% H_2O , 25-35% SO_2 , 15-20% N_2 and 10-15% CO_2 was treated at a gas flow rate of 55-60 m^3/hr , the reducing reaction temperature was 800-1000°C and the composition controlling reaction temperature was 700 to 850°C, thus obtaining results of an H_2S etc./ SO_2 ratio of 1.95-2.05, sulphur recovery yield

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of 93-97% and sulphur purity of at least 99.99%.

Example 3

5 Using an apparatus as shown in Figure 4 and a
coke oven gas having a composition of 55% H_2 ,
30% CH_4 , 7% CO , 1% O_2 and the balance N_2 and
10 CO_2 , a gas having a composition of 20-25% H_2O ,
20-25% SO_2 , 25-35% N_2 and 20-30% CO_2 was
treated to produce sulphur according to the process
as shown in Figure 3. Flow rate of the raw material
gas was $8\text{ m}^3/\text{hr}$, the reducing reaction temperature
was 700 to 800°C and the composition controlling
temperature was 600 to 700°C . The raw gas provided
15 for the Claus reaction was controlled to give an
 H_2S , etc./ SO_2 ratio of 1.95 to 2.05. Sulphur was
produced with a yield of 91 to 95% and a purity of at
least 99.99%.

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CLAIMS:

- 5 1. A process for treating a gas containing sulphur dioxide which comprises subjecting the gas to a reducing reaction and controlling by oxidation the composition of the gas to render the composition suitable for the Claus reaction.
- 10 2. A process according to claim 1, wherein a carbonaceous material is used as a reducing agent.
3. A process according to claim 1 or claim 2, wherein a reducing gas is used as a reducing agent.
- 15 4. A process according to any foregoing claim, wherein the reducing reaction performed at a temperature between 600 and 1000°C.
- 20 5. A process according to any foregoing claim, wherein the oxidation is performed at a temperature distribution of 500 to 900°C
- 25 6. A process according to any foregoing claim, whereing the said composition is controlled so as to give a ratio of $(H_2S + COS + 0.5CS_2)$ to XO_2 of 1.5 to 2.5.
- 30 7. A process according to any foregoing claim in which the said composition is controlled essentially by the supply of air.
- 35 8. An apparatus for treating a gas containing sulphur dioxide, comprising means for subjecting the gas to a reducing reaction and means for controlling the composition of the reduced gas by oxidation.

9. Apparatus according to claim 8, wherein the said means are provided in the same reaction vessel.

5 10. Apparatus according to claim 8, wherein the said reducing means comprise a reducing reactor and an oxidizing reactor adjacent to each other.

10 11. Apparatus according to claim 8, wherein the means for subjecting the gas to a reducing reaction is a moving bed reactor employing carbonaceous material and the said means for controlling the composition is a combustion chamber above the moving bed reactor.

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FIG. 1

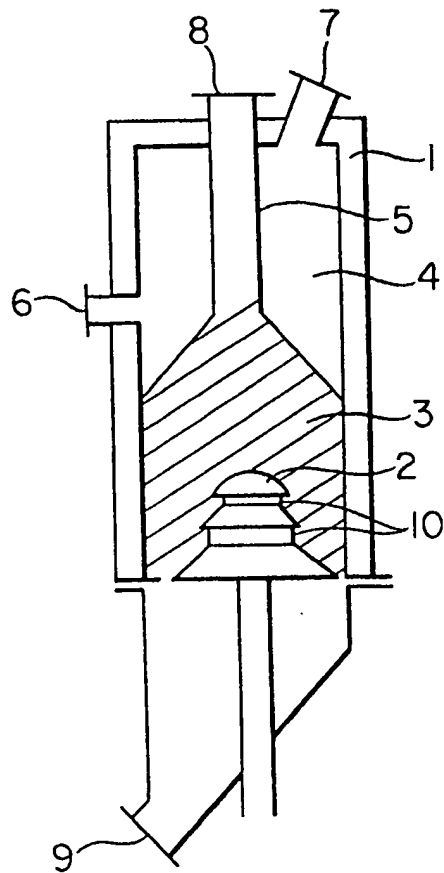
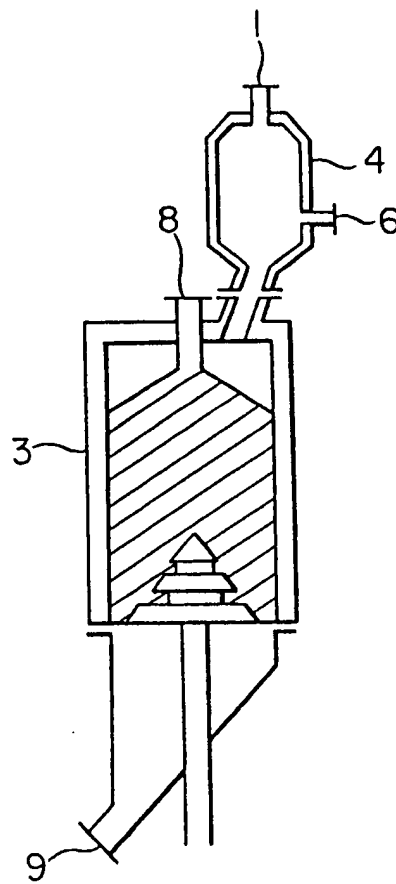


FIG. 2



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FIG. 3

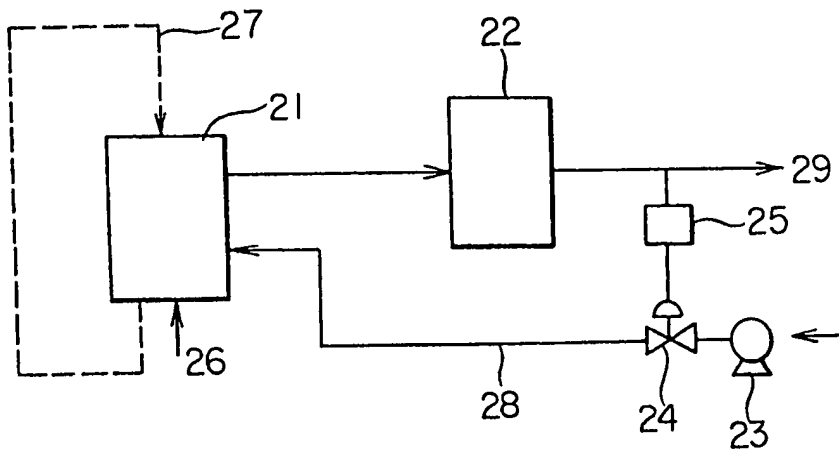


FIG. 4

